

TOPOLOGY OF DISORDERED GRAPHENE NETWORKS

Jacob W. Martin^{1*}, Carla de Tomas², Irene Suarez-Martinez², Markus Kraft¹ and Nigel Marks²

¹*Department of Chemical Engineering and Biotechnology, University of Cambridge, West Cambridge Site, Philippa Fawcett Drive, Cambridge CB3 0AS, United Kingdom*

²*Department of Physics and Astronomy and Curtin Institute for Computation, Curtin University, Perth 6012, Australia*

*Presenting author's e-mail: jwm50@cam.ac.uk

Introduction

The nanostructural topology of disordered graphene networks, otherwise known as isotropic or non-graphitising carbons, is an unsolved problem in carbon science. Rosalind Franklin suggested the first model whereby 3D connectivity between misaligned small graphitic domains was allowed for by crosslinks. Using electron micrographs, these crosslinks were found to be curved, leading others to develop ribbon-like models characterized by Gauss flat topologies. The discovery of fullerenes and the imaging of polygonalized structures in high-temperature carbons suggested a fullerene-like nanostructure characterized by Gauss positive curvature. Positive curvature, however, does not allow for connectivity in three dimensions and so nanostructures characterised by Gauss negative curvature were also suggested – Schwarzschild-like nanostructures. While these curved nanoforms are purely sp^2 , another topological element that has been proposed is Y/T-type junctions of sp^3 bonded carbon that allow for the termination of the graphene edge onto the face of another graphene fragment. While this could provide the porosity seen in some nanoporous carbons, the proportion of sp^3 carbon atoms would be too great. Evidence for all of these nanoforms has been found, but it is not obvious how these nanostructures are combined into a coherent model.

In this work, we approach the problem using a new mesh-based topological analysis of carbon nanostructural models generated using reactive molecular dynamics.

Methods

Periodic nanocarbon models were previously prepared using annealed molecular dynamics and provide close agreement to structural and electronic experimental measures.¹ A range of densities was chosen (0.5, 0.7, 0.9, 1.1 and 1.5 g/cc) that corresponds to the range over which the graphitic crystallites were found to self-assemble. For topological analysis we began by determining the positions of all rings. A triangular mesh was then constructed from the atoms within each ring and the vertex at the centre of each ring. This produced a unique decomposition of the carbon network into a discrete mesh whose topology could be probed. We made use of the angular defect about each vertex to determine the local and global curvature of each network.

Results and Discussion

Hexagonal arrangements of carbons atoms were found to dominate in all of the nanocarbon models. Within these networks were also a small proportion of non-hexagonal rings that arranged themselves into line dislocations (grain boundaries). Unlike planar polycrystalline graphene, these dislocations did not all form closed loops. Where these line dislocations ended without forming a

loop or where isolated non-hexagonal rings were present, non-zero local Gauss curvature was integrated. We found that counting non-hexagonal rings was unable to probe the global topology due to the networks being not purely sp^2 carbon. Instead, we used the angular defect. We found for all of the isotropic nanocarbon models an excess of negative angular defects indicating a net-negative Gauss curvature, which we propose is the characterising feature of isotropic carbons. **Figure 1** shows schematically the proposed *net-negative, fulleroid-like nanostructure*.

The non- sp^2 carbon atoms were then considered. At low densities we find Y/T-type and free edges dominate. With increasing density we also find that screw dislocations become dominant allowing for a winding up of the carbon sheets into a layered solid. This gives rise to the ribbons observed in glassy carbons and the cross-hatched regions, leading us to propose the *stacked, screwed, net-negative fulleroid-like nanostructure* for high density isotropic carbons. These results provide insights into porous carbons such as the integration of flexoelectricity,² glassy carbon ribbons and the graphitisability of carbon materials.

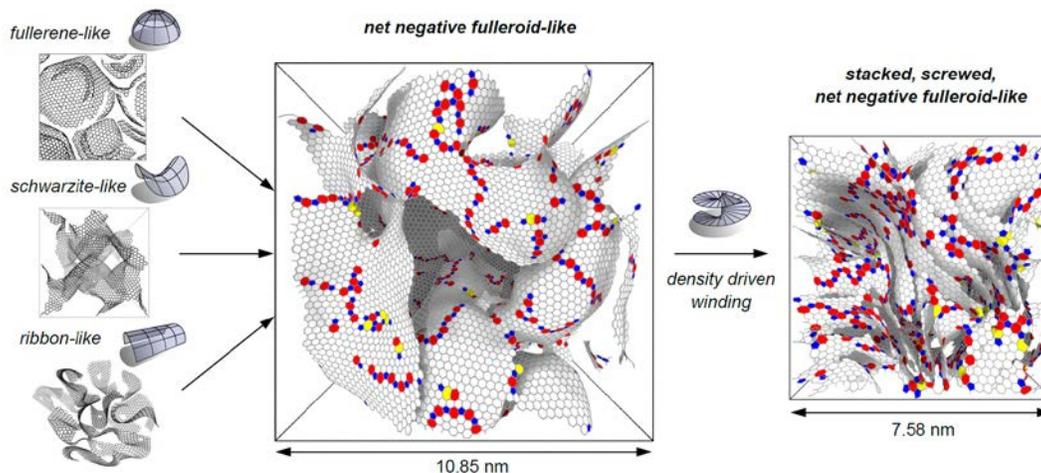


Figure 1. Schematic of the carbon nanoforms present in the models developed for disordered carbon.

Conclusions

Topological analysis of nanocarbon models prepared previously via reactive molecular dynamics reveals how different topological elements are integrated into carbon materials.

Acknowledgment

J. W. M. and M. K. acknowledge financial support from the National Research Foundation (NRF), Prime Minister's Office, Singapore, under its Campus for Research Excellence and Technological Enterprise programme as well as helpful discussions on differential topology with Dr Clive Wells, Hughes Hall, Cambridge.

References

1. De Tomas, C., Suarez-Martinez, I., Marks, N. A. (2018). Carbide-derived carbons for dense and tunable 3D graphene networks. *Applied Physics Letters* 112, 251907
2. Martin, J. W., Slavchov, R. I., Yapp, E. K. Y., Akroyd, J., Mosbach, S., Kraft, M. (2017). The polarization of polycyclic aromatic hydrocarbons curved by pentagon incorporation: the role of the flexoelectric dipole. *The Journal of Physical Chemistry C* 121, 27154-27163